Distinguishing Local Moment vs. Itinerant Ferromagnets: Dynamic Magnetic Susceptibility

Matthew D. Vannette,* Sergey L. Bud'ko, Paul C. Canfield, and Ruslan Prozorov

Ames Laboratory, Department of Physics and Astronomy,

Iowa State University, Ames, IA 50011

(Dated: 3 October 2007)

Radio frequency measurements of dynamic magnetic susceptibility of various ferromagnets show striking differences between local-moment (LFM) and weak itinerant (IFM) ferromagnetic systems. LFMs show a very sharp peak in susceptibility in the vicinity of the Curie temperature, T_C, that rapidly decreases in amplitude and shifts to higher temperature with the application of a weak dc bias field. In stark contrast, the generally accepted IFM systems show no peak, but rather a broad maximum well below T_C. The temperature of this maximum shifts to lower values and the amplitude is suppressed with an applied dc field.

Experimentally discriminating between local moment ferromagnetism (LFM) and itinerant ferromagnetism (IFM) is of theoretical and technological importance. From a theory standpoint, different models of ferromagnetism (FM) depend on the degree of localization of the moments (*i.e.* Heisenberg *vs.* Stoner). Technologically, itinerant ferromagnets are likely candidates for spintronic devices [1]. It is known that (fractional) size of the magnetic moment per ion alone is not a good indicator as the moment may be screened in a local system.

Typical susceptibility measurements are carried out below 100 kHz via amplitude-domain techniques. Frequency domain measurements are much more sensitive and can be realized over a wider frequency range. In the gigahertz regime, microwave cavity perturbation may be done. However, at these higher frequencies, samples are likely to be in the anomalous skin regime where the skin depth is less than the electron mean free path. Also, various resonant phenomena complicate the interpretation of the data. The radio-frequency (rf$\sim$10-100 MHz) band, on the other hand, largely avoids these problems, but it was difficult to access until

*corresponding author:vannette@iastate.edu

recently [2]. In this paper we use a highly stable, rf resonator to offer an experimental solution to the title problem.

I. EXPERIMENTAL DETAILS

The materials for this study were chosen based on their magnetic properties as determined by other conventional techniques. With one exception, all samples were single crystals grown as described in the respective references. The local moment systems chosen were $CeVSb_3$ [3], $CeAgSb_2$ [4], and GdPtIn [5]. $ZrZn_2$ [6] was chosen as a known itinerant system. Of the local moment systems, $CeAgSb_2$ ($T_C \approx 9.8$ K) has a rather small saturated moment of about 0.4 μ_B per Ce atom, which is of the order of $ZrZn_2$ ($M_{sat} \approx 0.2\mu_B$) [7]. Further, $CeAgSb_2$ is highly anisotropic, with the magnetic easy axis lying along the crystallographic c-axis. $CeVSb_3$ ($T_C \approx 4.5$ K) is also an anisotropic local moment system, however it has a larger saturated moment of about 4 μ_B. GdPtIn ($T_C \approx 68$ K) has a large moment, $M_{sat} \approx 7\mu_B$, with the easy axis along the hexagonal c-axis. $ZrZn_2$ ($T_C \approx 28$ K) is one of the prototypical itinerant ferromagnets exhibiting low anisotropy and the aforementioned small moment.

The real part of the radio frequency susceptibility, χ, was measured by using a sensitive tunnel diode resonator (TDR) technique. The design and capabilities of a TDR are discussed at length elsewhere [8–11]. Briefly, the resonance is driven by a tunnel diode that exhibits negative differential resistance when properly biased, and thus acts as a low current AC power source that compensates losses in a circuit. As a result, the circuit self-oscillates at the resonant frequency and the excitation field is very low (∼20 mOe). Properly designed and stabilized circuit allows one to measure changes in susceptibility on the order of a few parts per billion [2].

The sample to be studied is mounted on a sapphire rod with a small amount of low temperature grease and inserted into a small copper coil which acts as the inductor in a self-resonanting LC circuit. Changes in either the resistivity or bulk susceptibility of the sample induce changes in the resonant frequency of the LC circuit. It is straight-forward to show [12] that

$$\frac{\Delta f}{f_0} \approx -\frac{1}{2}\frac{V_s}{V_c}4\pi\chi. \tag{1}$$

where $\Delta f = f(H,T) - f_0$ is the change in resonant frequency due to the sample, f_0 is the empty coil resonant frequency, V_s is the volume of the sample and V_c is the volume of the coil. For insulating materials measured χ coincides with the static dM/dH, but for conductors normal skin effect has to be taken into account [2]. The temperature range for the TDRs used in this study is 0.4-150 K and a dc magnetic field provided by superconducting magnets in the range 0-90 kOe. The dc field is aligned with the coil inductor axis, thus the ac excitation field.

Lacking knowledge of f_0, it is very difficult to obtain an absolute value of χ from these measurements. Rather, the field and temperature dependencies in arbitrary units with an arbitrary zero are determined with extreme precision.

II. RESULTS

Upon cooling from above T_C, radio-frequency measurements of χ in local-moment systems exhibit a sharp peak in zero field (Fig. 1). Applying a dc bias field reduces the amplitude of the peak and shifts it to *higher* temperatures. The reduced amplitude is likely caused by the saturation of the moments by the external field leading to a smaller dM/dH. The thermal shift can be understood from a Weiss effective field argument wherein the applied field assists the molecular field in aligning the moments. Therefore, the maximum in χ signaling a crossover from the majority of the sample being uncorrelated to the majority being correlated shifts to higher temperatures. Similar zero field behavior is observed in measurements of specific heat, and the peak in χ observed here lies within the λ anomaly generally taken as the demarcation of the critical fluctuations regime. Critical scaling analysis can be done in this regime on our data [2].

Quite unexpectedly, the weak itinerant ferromagnet $ZrZn_2$ exhibits very different behavior (Fig. 2). On cooling through T_C there is no sharp increase in χ. Rather, there is a dramatic rise in susceptibility which passes through a broad maximum once the sample is in the ordered state. The application of a dc bias field suppresses the amplitude of this maximum, as one might guess. However, the temperature of the maximum shifts down, counter to what simple Brillioun variable ($\mu H/k_BT$) arguments would suggest. Since the response of the TDR includes contributions from both χ and ρ, a single crystal of $ZrZn_2$ was crushed with a mortar and pestle and the powder was mixed with a small amount of Stycast 1266 epoxy. The resulting pellet was composed of grains small enough that the skin depth at 10 MHz ($\sim$ 20 μm) was larger than the grain size ($\sim$ 10 μm). This effectively removed the

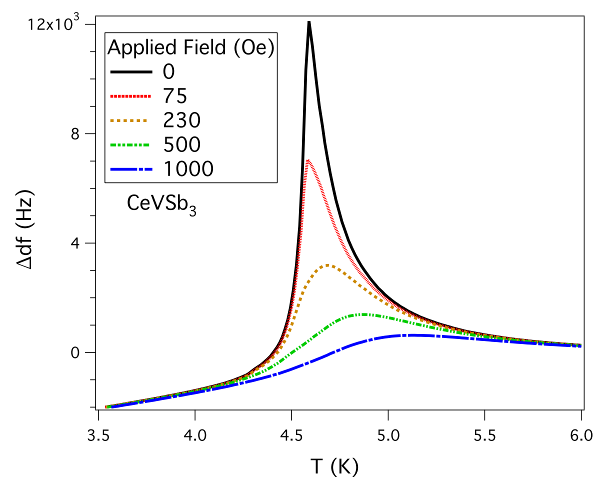

FIG. 1: (color online) Frequency shift ($\propto \chi$) of TDR resonance in single crystal $CeVSb_3$. The sharp peak is at $T_C \approx 4.5$ K.

resistivity component (a diamagnetic response) from the measurements, as shown in Fig 3. The field dependent temperature of the maximum below T_C is weakly quadratic.

There is a possibility of the $ZrZn_2$ powder being superparamagnetic, as the grains must be quite small to allow full penetration of the rf signal. This does not seem to be the case as a study of superparamagnetic EuS powder (to be published elsewhere) gives different results, particularly no maximum in susceptibility is seen below T_C.

A. Size of the Moment

Since $ZrZn_2$ has a small moment and $CeVSb_3$ does not, it is useful to compare two more LFM compounds: $CeAgSb_2$ ($M_{sat} \approx 0.4\mu_B$) and GdPtIn ($M_{sat} \approx 7\mu_B$). Figure 4 compares the zero field resonator response to a single crystal of $CeAgSb_2$ to that of GdPtIn. The behavior of both compounds is consistent with $CeVSb_3$ suggesting the peak behavior is indicative of local-moment magnetism irrespective of the size of the moment of the compound or the Curie temperature.

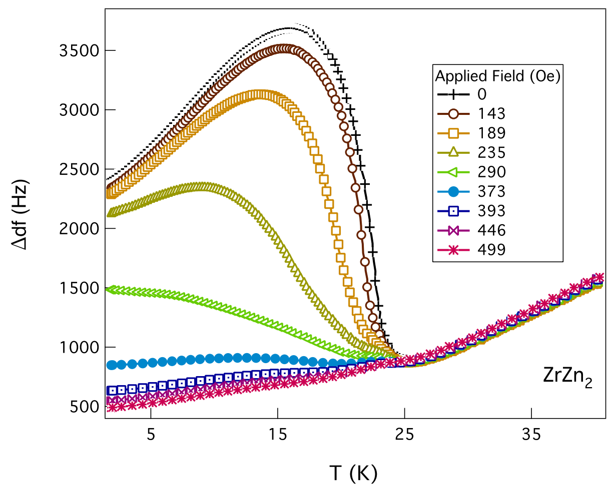

FIG. 2: (color online) Change in dynamic susceptibility of a single crystal of $ZrZn_2$ in various applied fields.

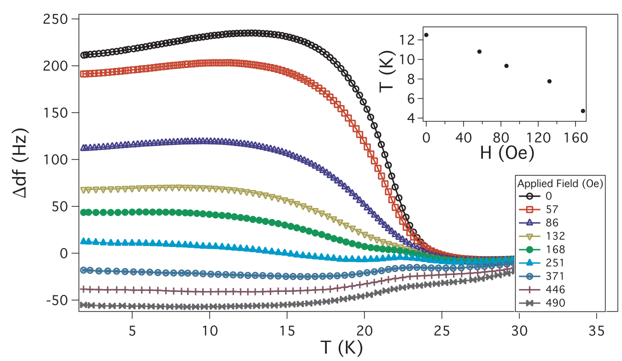

FIG. 3: (color online) Response of TDR to $ZrZn_2$ powder in epoxy. Inset: Scatter plot of temperature of maximum vs. applied field.

III. DISCUSSION

The rf susceptibility of LFMs may be understood in terms of Curie-Weiss arguments. In the vicinity of T_C the FM exchange interaction is assisted by the application of a fairly small field, susceptibility maxima shift to higher temperatures, and the divergence at T_C is suppressed [13].

However, the rf susceptibility of $ZrZn_2$ is not well understood. The temperature shift in the maximum below T_C is counter to expectations based on a Brillioun analysis of $\mu H/k_BT$.

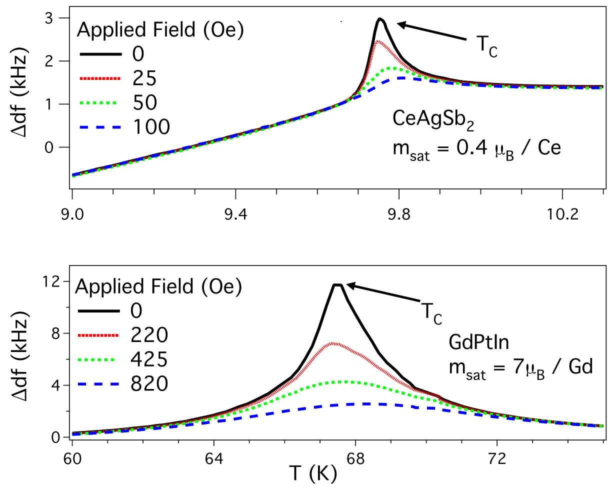

FIG. 4: (color online) Comparison of the zero field resonator response to the small moment $CeAgSb_2$ (top) with the large moment GdPtIn (bottom), both LFM compounds.

Beginning with the Stoner model for itinerant magnetism one expects $ZrZn_2$ to have a large density of states (DOS) at the Fermi energy [14] and a strong electron-electron interaction favoring parallel alignment of spins . The large DOS would imply a highly polarizable conduction band, as the energy cost of flipping a spin from the minority to the majority band is relatively small and offset by a decrease in the interaction energy [13]. This can account for the large χ in the ordered state, but not for the maximum. Ongoing work, including a study of Ni and Fe, will address this.

IV. CONCLUSIONS

It has been demonstrated that precise rf susceptibility measurements may be able to discriminate between LFM and IFM compounds. A study of more representative systems will provide the statistics needed to more fully support this conclusion.

V. ACKNOWLEDGMENTS

We thank Jörg Schmalian and Vladimir Antropov for useful discussions; Athena Sefat and Gerard Lapertot for providing some of the samples. Work at the Ames Laboratory was supported by the Department of Energy - Basic Energy Sciences under Contract No.

DE-AC02-07CH11358. R. P. acknowledges support from the Alfred P. Sloan foundation.

---

[1] J. Žutić, J. Fabian, and S. D. Sarma, Rev. Mod. Phys **76**, 323 (2004).

[2] M. D. Vannette, A. S. Sefat, S. Jia, S. A. Law, G. Lapertot, S. L. Bud'ko, P. C. Canfield, J. Schmalian, and R. Prozorov, doi:10.1016/j.jmmm.2007.06.018 (2007).

[3] A. S. Sefat, S. L. Bud'ko, and P. C. Canfield, *manuscript submitted.*

[4] K. D. Myers, S. L. Bud'ko, I. R. Fisher, Z. Islam, H. Kleinke, A. H. Lacerda, and P. C. Canfield, J. Mag. Mag. Mat. **205**, 27 (1999).

[5] E. Morosan, S. L. Bud'ko, and P. C. Canfield, Phys. Rev. B **72**, 014425 (2005).

[6] P. D. de Réotier, G. Lapertot, A. Yaouanc, P. C. M. Gubbens, S. Sakarya, and A. Amato, Phys. Lett. A **349**, 513 (2006).

[7] E. A. Yelland, S. J. C. Yates, O. Taylor, A. Griffiths, S. M. Hayden, and A. Carrington, Phys. Rev. B **72**, 184436 (2005).

[8] C. T. VanDegrift, Rev. Sci. Inst. **48**, 599 (1975).

[9] R. Prozorov, R. W. Giannetta, A. Carrington, and F. M. Araujo-Moreira, Phys. Rev. B **62**, 115 (2000).

[10] P. V. Parimi, H. Srikanth, M. Bailleul, S. Sridhar, R. Suryanarayanan, L. Pinsard, and A. Revcolevschi, arXiv:cond-mat/0007377v1 [cond-mat.str-el].

[11] R. Prozorov, M. D. Vannette, G. D. Samolyuk, S. A. Law, S. L. Bud'ko, and P. C. Canfield, Phys. Rev. B **75**, 014413 (2007).

[12] R. B. Clover and W. P. Wolf, Rev. Sci. Inst. **41**, 617 (1970).

[13] S. Blundell, *Magnetism in Condensed Matter* (Oxford University Press, 2001).

[14] J. Kübler, Phys. Rev. B **70**, 064427 (2004).